Ultratrace determination of neptunium-237 with resonance ionization mass spectrometry (RIMS)

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Introduction: After the decay of the fission products, ^{237}Np ($T_{1/2}$ = 2.14·10^6 a) will be a main contributor to the radiotoxicity of spent nuclear fuel. Therefore, the safety assessment of a nuclear waste repository must consider the geochemical properties of Np. Since the concentration of Np expected in the aquifer in case of a leakage of a nuclear waste repository is less than 10^{-10} mol/L, experiments dealing with the migration behaviour of Np require very sensitive analytical methods, e.g., resonance ionization mass spectrometry (RIMS).

In RIMS, the radiation from three tuneable lasers is used to excite and ionize sample atoms in subsequent steps. The uniqueness of optical transitions guarantees a high selectivity. Therefore, RIMS mass spectra are free of isobaric interferences. A prerequisite for the application of RIMS on Np is the development of a suitable excitation and ionization scheme. Numerous possible energy levels of ^{237}Np, which can be used for excitation and ionization, have been identified in a previous work[1].

Experimental: The laser system used for the generation of laser radiation consists of three titanium-sapphire (Ti:sa) lasers pumped simultaneously by a Nd:YAG laser at 532 nm. Blue laser radiation for the first excitation step is produced by frequency doubling in a BBO crystal. The individual laser beams are overlapped and transported into the atomic beam source. In the source region, Np atoms effusing from a heated filament are ionized by the laser light. The Np^+ laser ions are accelerated into a time-of-flight mass spectrometer and finally detected by a multichannel plate detector.

The filaments are prepared by electrodepostion of ^{237}Np onto a tantalum strip. In order to produce an atomic beam of Np, the strip is covered with a thin titanium layer acting as a reducing agent by magnetron sputtering. The thickness of the titanium layer depends on the sputtering time.

Several excitation schemes have been checked for their ionization efficiencies (Fig. 1). Also, the thickness of the sputtering layer has been varied. Typically, samples with 10^{10} atoms ^{237}Np have been used for efficiency measurements.

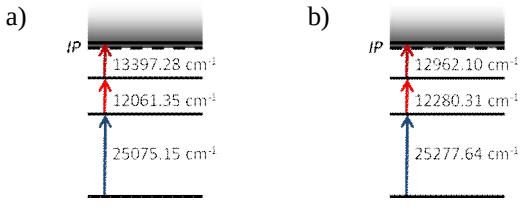

Fig.1: Two possible schemes for excitation and ionization of ^{237}Np.

Results: Due to the linewidth of 2-3 GHz of the Ti:sa lasers, a partial resolution of the hyperfine splitting (HFS) of ^{237}Np is possible (Fig. 2). Since the resolution of the HFS will lead to reduced ionization efficiency, energy levels with small HFS should be preferred. With the excitation scheme shown in figure 1b, a typical overall efficiency of approximately 3·10^{-7} was obtained.

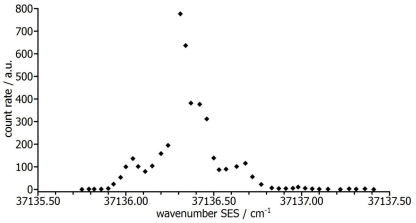

Fig. 2: Hyperfine splitting of the second excited state (SES) in the excitation scheme shown in Fig. 1a.

A slight improvement of the efficiency could be achieved by an increase of the sputtering time. After 120-180 min of sputtering, the filament is covered with a 1-1.2 µm thick Ti layer, resulting in an overall efficiency for the detection of ^{237}Np of 5·10^{-7} (fig. 3).

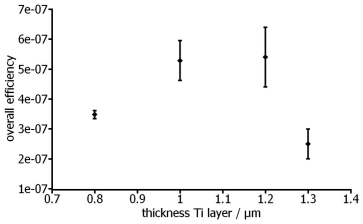

Fig. 3: Influence of the thickness of the Ti layer on the overall efficiency.

In all previous experiments, an optical fibre has been used for the transport of the laser light into the atomic beam source. However, the transmission efficiency of the fibre for NIR laser light is only about 60%. With the available laser powers, full saturation of the optical transitions was not possible for the second and third excitation step. After removal of the fibre and beam transport via high-reflectivity mirrors, the second excitation steps could be saturated. The overall efficiency increased to 2·10^{-6}.

References

[1] S. Raeder, N. Stöbener, T. Gottwald, G. Passler, T. Reich, N. Trautmann, K. Wendt: *Determination of a three-step excitation and ionization scheme for resonance ionization and ultratrace analysis of Np-237*, submitted to Spectrochimica Acta B

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